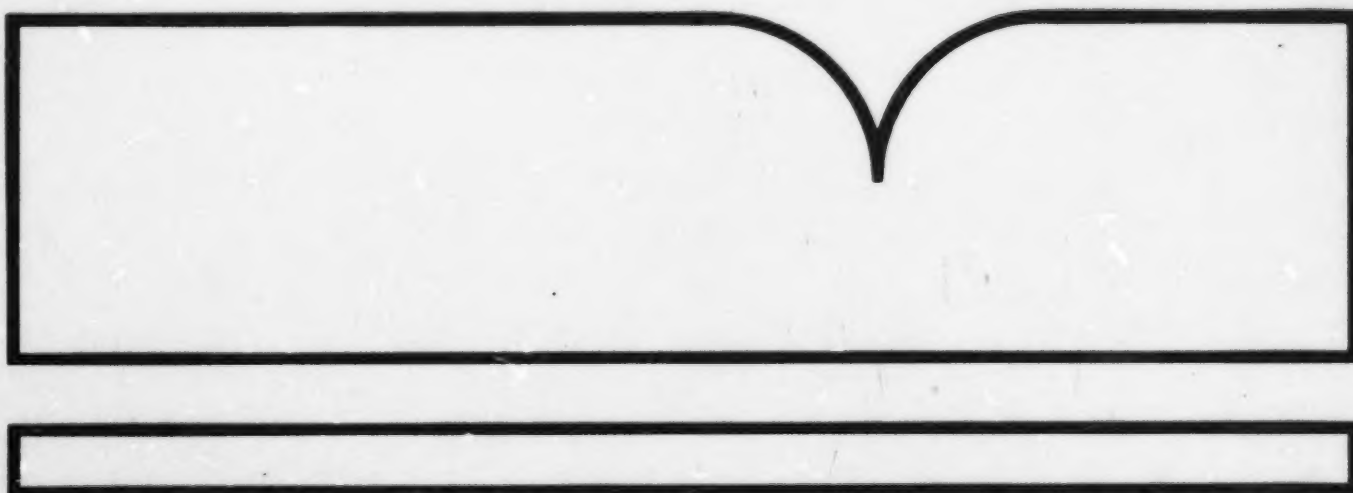


Water Treatment Model for
Pollutant Exposure Assessment

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WATER TREATMENT MODEL FOR POLLUTANT EXPOSURE ASSESSMENT

Key Words: Water Treatment, Toxic Substances, Exposure Assessment

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ABSTRACT

A mathematical model of potable water treatment for toxic substance removal for use in exposure assessment modeling has been developed. The sedimentation, coagulation-flocculation, filtration, aeration, chemical oxidation, and granular activated carbon adsorption treatment processes are modeled. The results of an example run of the model are presented.

INTRODUCTION

Under the Toxic Substance Control Act, the potential hazard to human health or to the environment of new chemicals must be evaluated before these compounds are marketed. An effective

technique in the process of assessing exposure is the use of mathematical models that predict the transport and transformation of toxic substances in the environment. These models can predict concentration levels in air, water, and food on the basis of the specific characteristics of the toxic substance and of the environment through which it passes. Figure 1 illustrates the interrelationships of these exposure routes to man.

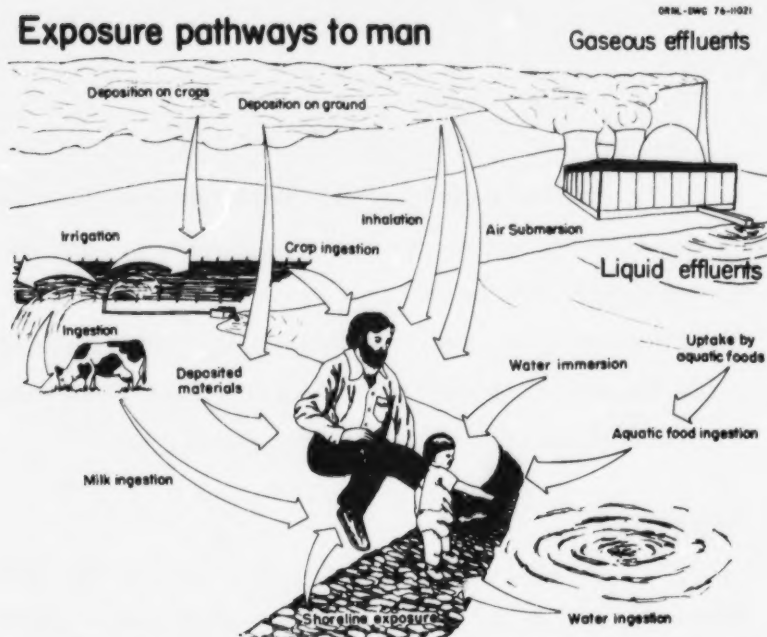


FIG. 1 EXPOSURE PATHWAYS TO MAN (Adapted from reference 2, courtesy of Oak Ridge National Laboratory, Health and Safety Research Division).

A potentially important exposure route for humans is the ingestion of chemicals in drinking water. Before reaching human receptors, these chemicals often must pass through municipal water plants where various treatment processes can act to change the pollutant concentration. A quantitative understanding of the transformations and removal processes in the treatment plant linked to information on the transport and transformation of toxic substances in the ambient source waters for the treatment plant should allow the development of mathematical models that can predict the concentration of a given chemical in drinking water actually at the tap. If the number and distribution of the population at risk is also known and the needed human health and toxicological data are available, the potential health hazard of exposure to the chemical can be assigned.

This paper describes a model of municipal water treatment processes for use in predicting the degree of removal of a toxic substance. Removal processes and transformation algorithms for this Water Treatment Processes (WTP) Model are, wherever possible, the same as those used in the model of transport and transformation processes in natural freshwater systems developed by SRI International¹ because predictions of chemical concentrations in the ambient source water using the SRI Model are used as input to the WTP Model.

DEVELOPMENT OF MODEL

The Water Treatment Processes Model accepts input data on influent streams, treatment processes configuration, and appropriate process rate constants for sedimentation, coagulation-flocculation, filtration, aeration, chemical oxidation, adsorption onto granular activated carbon columns, and chlorination. These treatment processes were selected for modeling because they are widely used in municipal plants in the United States. The program calls the appropriate process subroutine, i.e., one for each of the treatment processes listed above, in the sequence specified by the model user.

Sedimentation

Sedimentation, the oldest process for improving water quality for domestic use, uses subsidence and deposition to separate coagulated or suspended matter from the water. An effective means of water treatment, sedimentation was used in ancient Egypt, Carthage, and Rome³.

Hazen⁴ was the first to show that the floor area (and by analogy surface area) is the primary factor in determining how much suspended material may be removed by a sedimentation basin. Later investigators, including Camp⁵, have shown that "for any given discharge, the removal of particles is a function of the surface area and is independent of the depth of the basin; or, the removal is a function of the overflow rate and

for a given discharge is independent of the detention period." For the WTP Model, an empirical relationship was established between overflow rate and the removal efficiency of suspended solids, using the same approach as that of Smith⁶. Although few data are available for correlating overflow rate and suspended solids removal efficiency for water treatment, a fair amount of information has been published for wastewater treatment. Using data on removal efficiency of suspended solids versus overflow rate as tabulated and plotted by the American Society of Civil Engineers,⁷ the expression

$$\text{Sed}_{\text{rem}} = 0.76 * \text{Sed}_{\text{inf}} * e^{\left(\frac{-\text{OF}}{3247}\right)} \quad (1)$$

was derived, where the overflow rate (OF) is in gallons per day per square foot of surface area (gpd/ft²). If sorption of toxics to the suspended solids is taken into account and a mass balance is performed, the toxic substance concentration in the settling basin effluent can be calculated. Sorption is assumed to be completely reversible.

The fraction of the total amount of toxicant associated with particulate matter, defined as F, is determined by the following expression⁸,

$$F = \frac{1}{1 + w/(\text{Sed} * \text{CP})} \quad (2)$$

where w is the density of water. This expression can be solved for a range of sediment concentrations (Sed) and partition

coefficients (CP) to illustrate the relative importance of particulate matter as a toxicant transport medium⁸. As will be seen later, removal with particulates will be the major means of removal for those chemicals that tend to be sorbed by particulates.

Coagulation-Flocculation

Coagulation, the usual pretreatment before rapid sand filtration, involves the addition of chemicals to water, their uniform distribution through the water, and the buildup of a readily settleable floc. The process involves an initial high speed mix to ensure adequate, prompt dispersion of chemicals in the water being treated; an intermediate mix at lower speed, allowing flocculation and conditioning; and a period to allow sedimentation of the floc. It has been shown⁹ that the detention time is more important than the overflow rate in achieving flocculation. Coagulation-flocculation has been modeled for water treatment systems using either conventional coagulation-sedimentation or suspended solids contact clarification. For both cases, the formulations of Wanielista,^{10,11} which were empirically derived from data collected by Wanielista and from Kincaid,¹² were adopted for the WTP Model. Again the basic removal equation is an exponential expression:

$$Sed_{eff} = Sed_{inf} * e^K \quad (3)$$

The required data for the conventional coagulation-sedimentation process are the detention times for the flocculation basin (DTF) in minutes and the sedimentation basin (DTS) in hours. These detention times are used to determine K in the removal equation, where

$$K = (-0.062 - (0.07774 * DTF) ** 0.305 * DTS * 2.3).$$

Filtration

In recent years, water has been prepared for filtration in basins that combine mixing, flocculation, conditioning, sedimentation, and sludge removal in a single unit. Coagulation and flocculation take place in the presence of previously formed floc and excess solids are continually removed from the bottom. To model this suspended solids contact clarifier, the only additional parameter needed is the upflow velocity in the clarifier, because the upflow velocity determines the efficiency of the clarifier and the surface area determines the upflow velocity. K is then determined by the expression,

$$K = 0.891 * Q/V$$

where Q is the influent flow rate in MGD and V is the upflow velocity in ft/hr.

The most common device for treating municipal water supplies is the rapid sand filter, which removes nonsettleable

floc and impurities remaining after chemical coagulation and sedimentation of the raw water. Water passes downward through the filter by a combination of positive head and suction from the bottom. The action of the filter is extremely complex, consisting of straining, flocculation, and sedimentation. An exponential formulation adapted from Wanielista^{10,11} is used in the WTP Model:

$$\text{Sed}_{\text{eff}} = K1 * \text{Sed}_{\text{inf}}^{K3} e^{K2}, \quad (4)$$

where: $K = \frac{(SA * K2)}{Q}$

$$K1 = 0.83$$

$$K2 = 1.244 \text{ (MGD/ft}^2\text{)}$$

$$K3 = 0.81$$

$$SA = \text{surface area of filter (100 ft}^2\text{)}$$

$$Q = \text{flowrate (MGD).}$$

More exacting formulations can be developed but they require data that are not readily available.

The preceding processes predominantly remove particulate matter. Therefore, those toxic compounds that have a high partition coefficient will be preferentially removed with the particulate matter as quantified by equation 2. The following processes will address the chemicals remaining in solution.

Aeration

Aeration is normally used either to add oxygen to water or remove volatile substances from water. The WTP Model includes spray aeration and multiple tray aeration.

Spray aeration will increase volatilization of a compound, expose it to photodegradation during daylight hours and increase its potential for oxidation¹³.

The following expression is used^{1, 13, 14}.

$$\text{Conc}_{\text{eff}} = \text{Conc}_{\text{inf}} e^{-KT} \quad (5)$$

$$\text{where: } K = \frac{KV + KPD + (KOX * O_2)}{1 + CP * \text{Sed}_{\text{inf}} * 10^{-6}}$$

$$O_2 = \frac{475 - 2.65 * TDS}{33.5 + TP} * \frac{AP - WP}{760 - WP}$$

$$T = 1800^{-1} * CD * \sin \left\{ \phi * \sqrt{\frac{2h}{g}} \right\}$$

- KV = removal rate constant for volatilization, hr⁻¹
- KPD = removal rate constant for photodegradation, hr⁻¹
- KOX = removal rate constant for chemical oxidation, hr⁻¹
- CP = partition coefficient
- TDS = total dissolved solids, µg/l
- AP = atmospheric pressure, mm Hg
- WP = partial pressure of water vapor, mm Hg
- TP = temperature, °C
- CD = coefficient of discharge
- h = head on nozzle, ft
- g = 32.2 ft/sec²
- φ = angle of initial jet to horizontal

Scott¹⁵ developed a similar formulation for multiple tray aeration:

$$\text{Conc}_{\text{eff}} = \text{Conc}_{\text{inf}} * e^{-KT} \quad (6)$$

$$\text{where: } K = 0.0608 * \left(\frac{44}{MW} \right)^{1/2}$$

T = number of trays
MW = molecular weight of compound to be removed

Equation 6 only takes into account volatilization; 44 is the molecular weight of carbon dioxide, which was used as the reference compound because extensive data exist for it.

Photodecomposition would not be important here because, unlike spray aeration, multiple tray aerators are almost always enclosed. Oxidation is not included.

Chemical Oxidation

The purpose of oxidation in water treatment is to convert undesirable chemical species to species that are neither harmful or otherwise objectionable. Chemical oxidation processes were used as early as the eighteenth century to control taste and odor problems in drinking water. In those few early instances, simple oxygenation of public water supplies was employed to enhance oxidation of decomposed vegetation and animal matter to less offensive forms. This practice did not gain widespread use until the latter half of the nineteenth century, at which time oxygenation of public water supplies had become general practice¹⁵. As the overall benefits of oxidation, and the shortcomings of using atmospheric oxygen, became more clearly recognized, attempts were made to use stronger and more effective oxidants such as ozone, permanganate, chlorine dioxide, and chlorine¹⁶. In the WTP Model, an exponential decay function is again used. Values for the rate constants as a function of hydrogen ion activity (pH) must be determined. A

generalized temperature coefficient is used to account for increases in oxidation rate as temperature increases¹³.

$$\text{Conc}_{\text{eff}} = \text{Conc}_{\text{inf}} e^{-KT} \quad (7)$$

where:

$$K = (K_2) e^{(\text{slope (pH-7)})} \theta^{(T-20)} \text{Conc}_{\text{ox}}$$

K_2 = 2nd order chemical oxidation rate constant at pH 7 and 20°C

slope = slope of curve $\ln K$ vs. pH at 20°C

θ = temperature coefficient

T = temperature °C

Conc_{ox} = concentration of oxidant, mole/l

For this equation, data on the 2nd order oxidation rate constant at different pH values is needed so that both a range and a slope can be developed. Otherwise, measured values are required.

Adsorption onto Activated Carbon

Adsorption with activated carbon has traditionally been used to control taste and odor, but, with the advent of regulations for the control of trihalomethanes and other toxic organics, carbon adsorption by granular activated carbon columns will be more widely used. The granular carbon adsorption formulation in the WTP Model is based on the work of Kasha¹⁷ involving wastewater treatment systems. The carbon adsorption model is based on the premise that an effective lumped parameter model can be formulated. This formulation is possible if the competitive removal mechanisms of adsorption and bio-oxidation

are considered to be parallel rather than sequential mechanisms. On this basis, the following mass balance can be made.

$$E * VR * \frac{dC}{dt} = (C_{inf} * Q) - (C_{eff} * Q) - (E * VR * RXC) - (E * VR * RXS) \quad (8)$$

where: E = packed bed porosity
 Q = flowrate
 RXC = removal rate by adsorption
 RXS = removal rate by bio-oxidation
 VR = volume of contactor

This formulation is restrictive in that it does not allow for competitive adsorption between organics and does not account for byproducts resulting from transformations. The formulation, however, does allow for biomass sloughing that results in a net increase in suspended solids and particulate organics concentrations across the contactor.

Kinetic formulations developed for RXC and RXS¹⁷ are:

$$RXS = KFB (S - S_{IN}) \quad (9)$$

$$\text{where: } KFB = \frac{KF * NCPC * AB}{VR}$$

$$KF = \frac{JD * VS}{NSC^{2/3}}$$

$$JD = 1.34 * \left(\frac{RE}{1 - EPEX} \right)^{0.468}$$

$$NCPC = \frac{RHOPAC * VR}{RHOCF * VOLCAR * (1 - E)}$$

S = liquid concentration (M/L³)
 SIN = concentration within biomass (M/L³)
 KF = hydrodynamic film mass transfer coefficient (L/T)
 NCPC = number of carbon particles present
 AB = biofilm surface area per carbon particle (L²)
 VR = contactor volume = total volume of carbon times expansion factor (L³)
 JD = mass transfer factor
 VS = superficial velocity (L/T)
 NSC = Schmidt number
 RE = modified Reynolds number
 EPEX = expanded bed porosity
 RHOPAC = mass of carbon per unit contactor volume (M/L³)
 RHOCPC = mass of carbon per unit carbon volume (M/L³)
 VOLCAR = volume of one carbon particle (L³)
 E = packed bed porosity

All these parameters are either system-specific or easily calculated.

Film diffusion has been found to be rate-limiting in most applications.

$$RXC = KFA * A (S-S^*) \quad (10)$$

where:

$$KFA = \frac{1}{\frac{\delta L}{DL} + \frac{\delta B}{DB}}$$

S* = concentration within carbon averaged over entire carbon particle (M/L³)
 A = external mass transfer area (L²/L³)
 δL = thickness of hydrodynamic film
 δB = thickness of biofilm
 DL = free liquid diffusivity of solute (L²/T)
 DB = diffusivity of solute in biomass (L²/T)

Wen and Yu¹⁸ have established the following equation for activated carbon.

$$EPEX = \left(\frac{13.86 \text{ RHOL}^{.4} \text{ MJ}^{.6} \text{ US}^{1.4}}{\text{DCP}^{1.6} (\text{RHOC} - \text{RHOL})G} \right)^{.18} \quad (11)$$

where: EPEX = expanded bed porosity
 RHOL = density of solvent (M/L³)
 MU = viscosity of water (M/LT)
 US = superficial velocity (L/T)
 DCP = diameter of carbon particle (L)
 RHOBC = combined density of carbon and biomass (M/L³)
 G = gravitational constant (L/T²)

Having calculated the expanded bed porosity, equations 9 and 8 can now be exercised and solved for the steady state effluent concentration.

To accurately simulate the process, S must be estimated as a function of the fixed phase concentration, QE. The relationship between S and QE can be expressed as an isotherm such that S will be zero when QE is zero.
 An expression of the form

$$QE = b + m \cdot \log S$$

where: QE = equilibrium adsorbed phase
 concentration and b and m are
 constants

should give an acceptable prediction equation for S¹⁷.

Chlorination

Chlorination is the single most widely used water treatment process in the United States. Disinfection is its principal and most common objective, undoubtedly saving numerous lives by preventing epidemics of typhoid and other diseases.

Chlorination, however, may also cause formation of toxic

chlorinated compounds or may oxidize chemicals in the raw water to more toxic forms. Whether a given compound is changed depends primarily on its oxidation state, with the higher states less likely to change. Insufficient data are available to permit the development of a generalized chlorination model at this time. Rather, each new compound will need to be considered individually for predictive purposes. The extent of any change due to chlorination will depend upon the chlorine dosage, toxicant concentration, and time of contact. Moore et al.¹⁹ developed a regression model relating chlorine dosage to the chloroform concentration in the distribution system. If enough data of this type for a specific chemical are collected, it would be relatively simple to develop a general regression model for that chemical.

RESULTS AND DISCUSSION

The primary objective was to develop a model of water treatment processes that provides a means of predicting the quantity of a specified toxicant in the finished potable water. When such a model is linked to an aquatic fate and transport model, a source identification and loading model, and a model for estimating the population at risk, the actual amount of toxicant reaching a population for consumption in their drinking water can be predicted. SRI International has developed an aquatic fate and transport computer model and

supporting laboratory procedures to measure the sorption partition coefficient and the rate constants for volatilization, oxidation, hydrolysis, photolysis, and microbial transformations, under conditions that are extrapolatable to environmental conditions¹. The SRI researchers applied these procedures to 11 toxic chemicals of environmental concern, and used the results with their aquatic fate and transport model to predict the pathways for these chemicals in ponds, lakes and rivers²⁰. Their descriptions, coefficients and constants are also used whenever possible as input to the WTP Model.

Table 1 shows the partition coefficients and rate constants for volatilization (KV), photodegradation (KPD), and oxidation (KOX) used for the 11 toxic chemicals modeled by SRI

TABLE 1
Rate Constants of 11 Selected Chemicals

Chemical	CP ($\times 10^{-3}$)	KV ($\times 10^4 \text{ hr}^{-1}$)	KPD ($\times 10^2 \text{ hr}^{-1}$)	KOX ($\times 10^5 \text{ hr}^{-1}$)
p-Cresol	.01	40	.05	7
Quinoline	.01	1	.06	7
Methyl Parathion	.05	.02	.2	.35
Benzo thiophene	.05	150	.06	.70
9H-Carbazole	.2	.07	11	290
Benzoquinoline	1.4	.08	25	.01
Dibenzothiophene	1.4	50	.18	.70
7H-Dibenzocarbazole	20	.47	69	100
Benzoanthracene	25	.07	3.4	1800
Benzopyrene	50	50	23	200
Mirex	300	14	.009	7

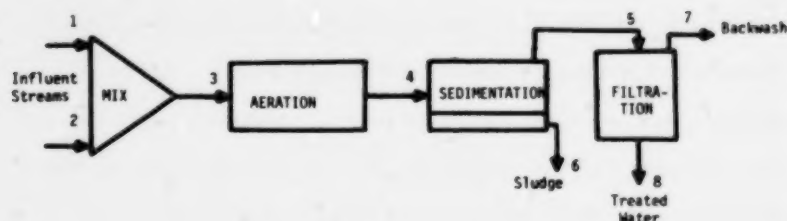


FIG. 2 EXAMPLE WATER TREATMENT PROCESS TRAIN

International. As an example, if the WTP Model is run using the water treatment process train shown in Figure 2 and the SRI partition coefficients and rate constants, the removals shown in Table 2 are obtained for these 11 compounds.

These results clearly illustrate the effect that the partition coefficient (CP) of a toxicant has upon its mechanism of removal in water treatment. Toxic chemicals that have a high partition coefficient, and hence a high affinity for sorption to

TABLE 2
An Example of Predicted Removals Using WTP Model for
11 Selected Chemicals

CP ($\times 10^{-3}$)	% Removal With Sediment	K's ($\times 10^2 \text{ hr}^{-1}$)	% Removal in Water Phase	% Removal (Total)
.01	.1	.05	9.1	9.2
.01	.1	.077	9.2	9.3
.05	.7	.2	10.1	10.8
.05	.7	1.5	11.8	12.5
.2	2.9	11	58.4	61.3
1.4	16.7	25	31.8	48.5
1.4	49.6	.68	3.9	53.5
20	63.0	70	17.0	80.0
25	67.2	5.2	2.6	69.8
50	76.4	24	2.7	79.1
300	80.9	.14	0.2	81.1

particulate matter, will preferentially be removed by those processes that tend to remove particulate matter. Hence, a chemical such as mirex, which is almost completely sorbed to particulates, will be removed at or near the same rate as the particulates. Thus, processes such as sedimentation, coagulation-flocculation, and filtration will be highly effective in its removal from the product water. A chemical that has a low partition coefficient such as quinoline, however, will be essentially unaffected by those processes that remove particulate material. Consequently, chemicals that do not readily sorb to particulates are not removed to any appreciable extent unless they also volatilize or undergo some other transformation; in which case, aeration, chemical oxidation, or carbon adsorption may be effective in reducing the toxicant's concentration in the finished water. The detention time of water through the water treatment system is commonly on the order of one day. Therefore, it should be noted that compounds that have halflives significantly greater than the detention time of the system will not be degraded or volatilized during treatment to any great extent because, by definition, the halflife of a substance is that time taken for one half the material present to undergo change. In general for significant degradation to occur, the halflife must be on the order of one third of the detention time of the process or system under consideration.

Thus, if the influent to the water treatment plant diagrammed in Fig. 2 contained 60 $\mu\text{g/l}$ of mirex and 6 g/l of quinoline (the approximate solubility limits), then it would be predicted by the WTP Model that the effluent concentrations would be 3.5 $\mu\text{g/l}$ and 5.4 g/l, respectively.

Summary

The WTP Model calls subroutines for each of the treatment processes being modeled, an output subroutine and several other supporting subroutines. The process subroutines presently include sedimentation, coagulation-flocculation (two versions), filtration, aeration (two versions), chemical oxidation and granular activated carbon adsorption. The chlorination subroutine needs more experimental data upon which to base the modeling effort, and in all probability chemicals will need to be modeled on an individual basis. Any specific process can be similarly added for which enough experimental information is available upon which to construct a model algorithm. The WTP Model provides a means of predicting the quantity of a toxic in tap water following treatment. When linked with an aquatic fate and transport model, a source identification and loading model, and a model for estimating the population at risk, the WTP Model can be used to predict the amount of a toxicant actually reaching a population for consumption by the drinking water route.

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